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The wreckage of the oil tanker 'Erika'—human health risk assessment of beach cleaning, sunbathing and swimming☆

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Dedicated to the late Philip Chambers

Abstract

In December 1999 the oil tanker 'Erika', carrying approximately 30 tons of heavy fuel oil, wrecked before the coast of Brittany (France), polluting the local beaches and rocks over a distance of some 500 km. Also numerous birds were affected. During the first months of 2000 the coastal area and many birds were cleaned. The health risk for people involved in these cleaning activities and for tourists was evaluated with emphasis on the carcinogenic properties of this oil. The outcome indicates that the risks were limited to people who had been in bare-handed contact with the oil. Firstly they had an increased risk for developing skin irritation and dermatitis, however, these effects are in general reversible. Secondly they had an increased risk for developing skin tumours, but since the dermal contacts with the oil were of relative short duration, this risk is considered to be very limited.

Keywords: 'Erika'; Heavy fuel oil spill; Pollution; Human health; Risk assessment

Abbreviations: bw, body weight; HC, hydrocarbon; *K*ow, octanol/water partition coefficient; LOAEL, lowest observed adverse effect level; MRL, minimal risk level (as defined by ATSDR); NOAEL, no observed adverse effect level; PAH, polycyclic aromatic hydrocarbon; RfC, reference concentration (as defined by US-EPA); RfD, reference dose (as defined by US-EPA); TCA, tolerable concentration in air; TDI, tolerable daily intake; TPH, total petroleum hydrocarbon.

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1. Introduction

On Sunday December 12, 1999, the 30 000 ton Maltese oil tanker 'Erika' wrecked on the Atlantic Ocean about 65 km south of Penmarch (Brittany, France), causing a spillage of approximately 10 000 tons of oil, contaminating the French coast and its small islands from Southern Brittany to

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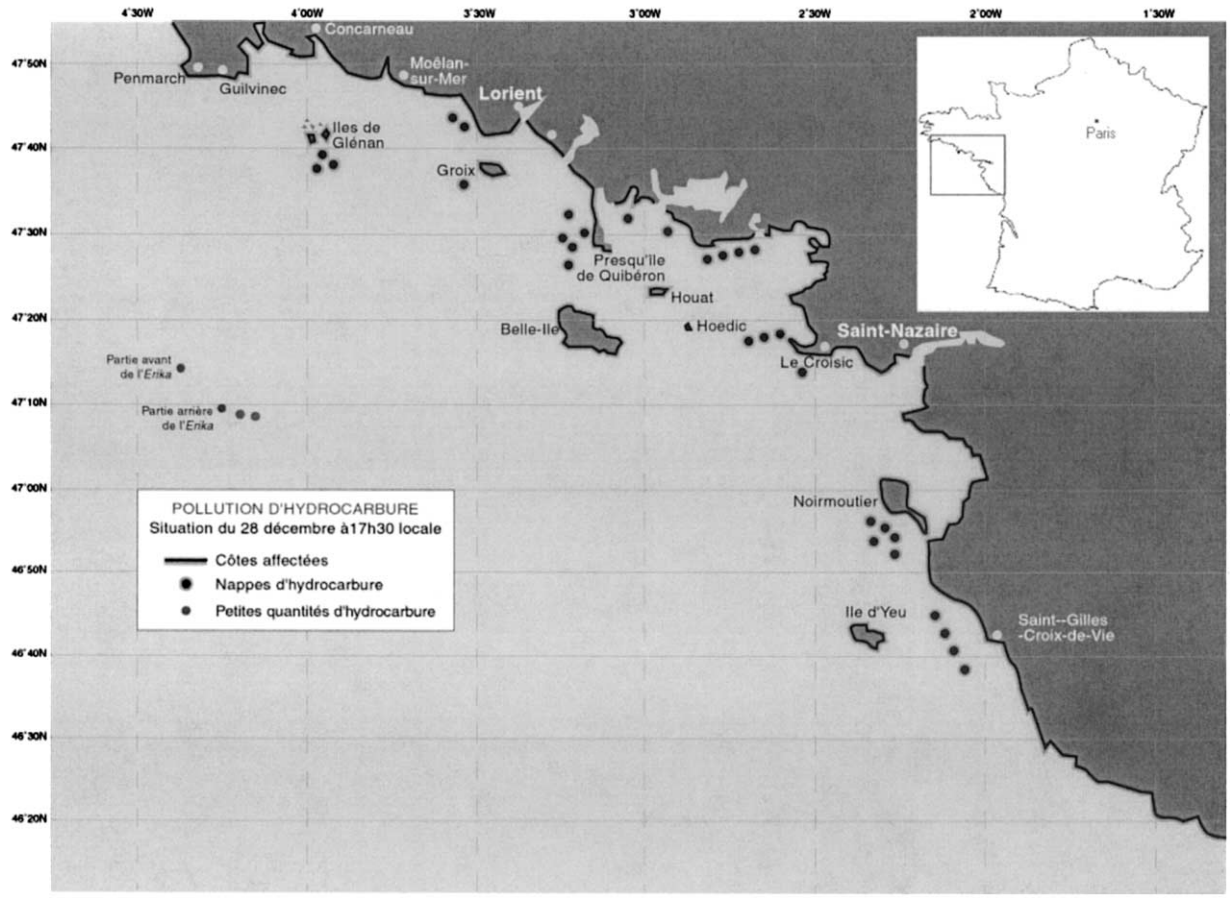

Fig. 1. Location of the 'Erika' and the extent of the pollution on the coast of southern Brittany (France), on December 28, 1999. Source: internet homepage of the French Ministry of the Environment, February 2000 (www.environnement.gouv.fr).

the Vendée over about 400–500 km (Fig. 1). For a large part, cleaning of the polluted coast had to be done by hand due to the rough and rocky nature of the coast; in addition numerous oil-contaminated birds were cleaned.

Early February 2000, the French Ministry of the Environment (Ministère de l'Aménagement du Territoire et de l'Environnement, Direction de la Prévention et des Risques, Paris) asked the Dutch National Institute of Public Health and the Environment (RIVM) for an independent risk assessment for humans involved in the beach-cleaning operations, at mid-April followed by a similar request dealing with tourist activities on these beaches.

2. Oil spillage data

2.1. Introduction

A number of analyses[1] identified the spilled oil as 'fuel oil # 6' (CAS no. 68553-00-4), also

[1] A large number of local samples were analysed by the Centre de Documentation de Recherche et d'Experimentations sur les Pollutions Accidentelles des Eaux (CEDRE), Brest, France; some samples taken at sea by the 'Arka' (a Dutch ship designed for the containment of oil spills) were analysed by the Institute for Inland Water Management and Wastewater Treatment (RIZA), Lelystad, The Netherlands; the National Institute of Public Health and the Environment (RIVM) analysed some samples taken from polluted birds that were transported to Dutch bird rehabilitation centres for cleaning.

known as 'bunker oil C' (Boudet et al., 2000). Fuel oil #6 is one of the so-called 'heavy fuel oils' which in turn constitute part of the 'residual oils'.

2.2. General information[2]

Heavy fuel oils are composed of a mixture of petroleum distillate HCs. Fuel oil #6 is the highest boiling fraction of the heavy distillates from petroleum; its boiling point is >200 °C (and generally <400 °C). Fuel oils #4, #5, and #6 are known as 'residual oils', because they are produced from distillation residues from refinery processing. To produce fuels that can be conveniently handled and stored, the high viscosity residue components are normally blended with gas oils or similar low viscosity fractions; also catalytically cracked cycle oils are common fuel oil diluents. As a result, the composition of residual fuel oils can vary widely. The most viscous residual fuel oils, #5 and #6, usually must be preheated before being burned.

Heavy fuel oils are complex and variable mixtures of relatively high molecular weight compounds, and difficult to characterise in any detail. Generally these oils include asphaltenes (highly polar aromatic compounds with molecular weight 2000–5000), aromatics (including polar and naphthalene aromatics), saturated HCs, and heteromolecules containing sulphur, oxygen, nitrogen and metals. Fuel oil #6 contains about 15% paraffins[3], 45% naphtenes[4], 25% aromatics (most residual fuel oils are likely to contain ≥5% four- to six-ring condensed aromatic HCs), and 15% non-HC compounds; the HC compounds typically having carbon numbers from C_{20} to C_{50}. Di- and trimethylnaphthalenes are important components of fuel oil #6. It also contains organometallic compounds from their presence in the original crude oil; the most important of these are vanadium, aluminium and nickel. Oil-soluble compounds of calcium, cerium, iron or manganese may be added to residual fuel oils to improve combustion; concentrations vary, but typically range between 50 and 300 mg of the active ingredient per kilogram of oil. According to the literature, the $\log K_{ow}$ of fuel oil #6 varies between 2.7 and 6.0 (CONCAWE, 1998) and/or 3.3 and 7.1 (Irwin et al., 1997), indicating a high bioaccumulation potential of at least part of its components.

Fuel oil #6 has a low evaporation or dissolution potential. As such it is highly persistent, with the potential for long-term (in the order of magnitude of several years) sediment and coastal rocks contamination.

The density of fuel oil #6 is generally slightly less than 1 (a typical fuel oil #6 has a density of 971 kg/m^3 at 22 ± 2 °C), which may increase to values ≥1 upon weathering, causing risk of sinking (freshwater density is approximately 1000, and seawater density is approximately 1024 kg/m^3), thereby impacting benthic and sessile organisms. Emulsions formed by fuel oil #6 are very stable.

During a spill, the high viscosity of fuel oil #6 often leads to the formation of 'pancake'-like tar globs when the temperature of the water is lower than the pour point of the oil. These semi-solid, tar-like oils have low penetrating ability, and are difficult to remove from contaminated surfaces.

Weathering of this fuel oil is very slow. In 28-day laboratory studies with crude and refined oils, fuel oil #6 was the least degraded: 11% compared to 51–82% for crudes, due to its higher proportion of high molecular weight aromatics.

Some general physical–chemical parameters of fuel oil #6 are summarised in Table 1.

2.3. Pollution data

In the last week of December 1999 the Centre de Documentation de Recherche et d'Experimentations sur les Pollutions Accidentelles des Eaux (CEDRE; Brest) analysed the oil and characterised it as fuel oil #6 (according to US, UK and international nomenclature; French nomenclature classifies this type of residual oil as 'fuel oil #2');

[2] The introductory information in this section is summarised from ATSDR (1995a, 1999), IARC (1989), Irwin et al. (1997), CONCAWE, 1998. To increase readability, references in the text are limited to the absolute minimum.

[3] *n*-Alkanes and branched alkanes.

[4] Cyclic alkanes.

Table 1
General physical–chemical data of fuel oil # 6

Fuel oil # 6 (Bunker C)	CAS no. 68553-00-4
Approximate composition (% w/w)	Alkanes 15% Cycloalkanes 45% Aromatics 25% Non-hydrocarbon compounds 15%
Boiling point	Approximately 400 °C
Melting point	Approximately −46 °C
Pour point[a]	−4 to +15 °C
Viscosity (dynamic)	48000 cp (at 10 °C)
Density	0.96–0.98 kg/m^3 (at ambient temperature)
Water solubility	≤5 mg/l
log K_{ow}	2.7–7.1
Flash point	66–132 °C
Ignition temperature	407 °C

All data are at 0% weathering, i.e. fresh fuel oil # 6.
[a] The 'pour point' is the lowest temperature at which an oil sample is observed to flow when cooled under prescribed conditions.

the results of these analyses are summarised in Table 2 (Boudet et al., 2000). In addition, the PAHs present in this oil were analysed (limited to the 16 so-called 'EPA-PAHs', i.e. the PAHs recommended for analysis by the US Environmental Protection Agency in pollution situations); the results of these analyses are listed in Table 3 (Boudet et al., 2000).

Table 2
Chemical analysis of the fuel oil # 6 cargo of the Erika

Elementary analysis (% w/w)	Fresh oil	
Carbon	88.20	
Hydrogen	10.42	
Oxygen	1.66	
Nitrogen	0.39	
Sulphur	2.50	
Compound classes (% w/w)	Initial	Weathered
Saturated hydrocarbons	21.8 ± 1.1	22.1 ± 1.1
Aromatic hydrocarbons	42.3 ± 1.1	42.2 ± 1.1
Resins	31.3 ± 1.1	31.1 ± 1.1
Asphaltenes[a]	4.6 ± 0.5	4.5 ± 0.5

[a] Highly polar aromatics with molecular weights 2000–5000.

Operations aiming at cleaning the polluted coastal areas and cleaning oil-contaminated birds were started very quickly after the wreckage. Due to the rather wild nature of the contaminated area, a large part of the cleaning had to be done manually. Birds were cleaned bare-handed, using various detergents. The protective clothing used in these activities was cleaned on a daily basis using high-pressure water devices.

In view of the cleaning activities and the potential health risk involved, some actual exposure measurements were performed in the contaminated area by mid-February 2000 (Boudet et al., 2000). Atmospheric analyses indicated the presence of 5.2 and 5.9 ppm (v/v) of volatile organic compounds; assuming an oil density of 0.97 (Table 1) this equals 3920 and 4450 μg/m^3, respectively. In laboratory simulation tests this vapour was shown to contain benzene, toluene, xylenes and naphthalene: see Table 4. Other analyses were not performed.

Cleaning the polluted protective clothing created a humid and PAH-rich atmosphere in which in two separate analyses 23 and 33 ng benzo[*a*]pyrene equivalents (based on the 16 EPA-PAHs), and 620 and 680 ng naphthalene per m^3 were found (see Table 5). Other analyses were not performed.

For the current risk assessment further analytical–chemical and/or exposure data were not available.

3. Toxicology[5]

3.1. Introduction

The heavy fuel oils are less acutely toxic relative to other oil types. The carcinogenicity of petroleum products including fuel oils, however, is a major cause for concern. It is generally accepted

[5] The information in this section is summarised from ATSDR (1995a,b, 1999), IARC (1987, 1989), IPCS (1982, 1998), TPHCWG (1997a,b), CONCAWE (1998), Baars et al. (2001). To increase readability, references in the text are limited to the absolute minimum.

Table 3
Analysis of some aromates and polycyclic aromates (16 EPA-PAHs) present in the oil spill

Compound (mg/kg)	Fresh oil[a]	Beach sample 1[b]	Beach sample 2[b]
Naphthalene	534	96	21
Acenaphtylene	1	5	5
Acenaphtene	126	27	29
Fluorene	141	33	38
Phenanthrene	535	136	169
Anthracene	94	20	22
Fluoranthene	49	11	12
Pyrene	279	91	115
Benzo[*a*]anthracene	298	72	105
Chrysene	508	159	231
Benzo[*b*]fluoranthene	39	22	31
Benzo[*k*]fluoranthene	19		
Benzo[*a*]pyrene	153	36	70
Dibenzo[*a*,*h*]anthracene	21	12	18
Benzo[*g*,*h*,*i*]perylene	42	17	20
Indeno[*c*,*d*]pyrene	11	10	18
Total	2850	747	904
Benzo[*a*]pyrene equivalents[c]	217	60	107
Benzene	–	< 0.2	0.5
Toluene	–	0.7	1.8
Xylenes	–	2.7	9.5

[a] Oil sample of TOTALFINA, taken from oil freshly arrived on the beach in week 4, 2000, ref. DA589/No1749 (Boudet et al., 2000).

[b] Results according to Boudet et al. (2000), analysed week 7, 2000. Sample 1 taken from oil that arrived on the beach Feb. 15 and was sampled the same day (ref. 00CS17), sample 2 taken from oil that arrived some days earlier and was also sampled Feb. 15 (ref. 00CS18).

[c] Benzo[*a*]pyrene equivalents calculated according to Baars et al. (2001).

that the PAHs, which are present in a number of fuel oils, are responsible for the carcinogenic effects following exposure to these oils.

General toxic risks can be evaluated applying the threshold approach (because toxicity only becomes manifest above the NOAEL); for carcinogens with a genotoxic mode of action, however, such a threshold is assumed not to exist, and consequently in the evaluation of these carcinogens the non-threshold linear extrapolation approach is generally applied.

The toxicology and carcinogenicity of some mixtures such as diesel fuel, fuel oils and gasoline, and a number of individual constituents has been evaluated by several organisations, including the US Agency for Toxic Substances and Disease Registry (ATSDR, 1995a,b, 1999), the International Agency for Research on Cancer (IARC, 1987, 1989), and the International Programme on Chemical Safety (IPCS, 1982, 1998). The toxicological information of many constituents is limited. In 1997 the US Total Petroleum Hydrocarbon Criteria Working Group (TPHCWG), examining information on 254 chemicals in the C_3–C_{26} range, identified approxi-

Table 4
Atmospheric samples on the oil-polluted beach

Compound (μg/m^3)[a]	Sample 1	Sample 2
Volatile organic compounds	3920	4450
Naphthalene[b]	4.22	4.62
Benzene[b]	124	96
Toluene[b]	489	576
Xylenes[b]	1687	1995

[a] Data taken from Boudet et al. (2000).

[b] Values obtained in laboratory simulations (dynamic test chamber for naphthalene, static test chamber for benzene, toluene and the xylenes), and used as such for risk estimation.

Table 5
Atmospheric samples during cleaning of clothes (using high pressure water)

Compound ($\mu g/m^3$)[a]	Sample 1	Sample 2
Benzo[*a*]pyrene equivalents	0.023	0.033
Naphthalene	0.62	0.68

[a] Data taken from Boudet et al. (2000).

mately 65 compounds for which at least some useful toxicological information was available (TPHCWG, 1997a,b), which was essentially confirmed and adopted by the ATSDR (1999). In addition, the Oil Companies' European Organisation for Environment, Health and Safety (CONCAWE) has started to publish a series of reports summarising the available information on toxicity etc. of a number of principal oil products; the 1998 report deals with heavy fuel oils (CONCAWE, 1998). RIVM evaluated TPHs and PAHs in 2000, estimating TDIs, TCAs, and carcinogenic risks for the various TPH compound classes and a number of individual PAHs (Baars et al., 2001).

3.2. Acute/subacute/subchronic toxicity

In general, petroleum distillates are rather poorly absorbed from the gastrointestinal tract and do not cause appreciable systemic toxicity by ingestion unless inhalation occurs, in which case primary effects include pulmonary damage and transient depression or excitation of the central nervous system. In addition, inhalation exposure to volatile petroleum HCs may result in cardiac arrhythmias. Case reports of renal and haematological effects have also been recorded from acute high exposure. Long-term exposure of humans living up to 17 years nearby an oil processing plant in a heavily contaminated area was reported to have resulted in neurophysiological and neurological impairment.

Heavy fuel oils containing relatively high amounts of sulphur compounds may release H_2S, which is highly toxic, causing effects such as eye irritation, nervousness, nausea, headache, insomnia and, in severe cases, unconsciousness and death.

Dermal effects from short-term exposure to heavy fuel oils include irritant and defatting effects, resulting in primary irritation, dryness, cracking, oil acne, dermatitis, hyperkeratosis and photosensitivity. Exposure to heavy fuel oils may lead to severe eye irritation, in contrast to fuel oils with lower boiling points, which are more irritating to the skin. Fuel oil # 6 demonstrated mild dermal sensitising potential. Exact data regarding dermal exposure of humans are extremely scarce. Data summarised by ATSDR (1995a) indicate that light types of fuel oil (in particular diesel fuel) are absorbed via the skin of humans, but quantitative data are not available. There are no data for other fuel oils.

LD_{50} values for heavy fuel oils range from 4700 mg/kg bw to 17500 mg/kg bw if orally administered to rats, and from >2000 to >5000 mg/kg bw if dermally administered to rabbits.

The scarce data available on effects upon subacute/subchronic exposure are restricted to inhalation studies with experimental animals and indicate mainly nephrotoxic and pulmonary effects, but liver effects have also been reported. It must be noted, however, that the toxic effects in male rat kidney observed with various HCs are the result of a complex accumulation process that starts with the interaction of HC metabolites and alpha-2u-globulin. The accumulation causes tubular cell damage and increased cellular proliferation, which enhances the probability of tumour development. When alpha-2u-globulin is not produced in substantial amounts (such as in female rats, mice, or other animal species including man), neither the nephrotoxicity nor the subsequent carcinogenesis occurs.

3.3. Developmental and reproductive toxicity

Heavy fuel oils showed maternal and foetal toxic effects in rats (19 days dermal exposure starting at day 0 of gestation) at doses of 8 and 30 mg oil per kg bw per day, respectively (LOAELs).

3.4. Carcinogenicity

Human epidemiological studies have demonstrated the association of petroleum HC expo-

sures with various adverse health outcomes. Of these, carcinogenicity is of particular importance; in general this is mainly attributed to the presence of PAHs in the various petroleum HCs. Inhalation and/or dermal exposure to mineral oils that have been used in a variety of occupations, including mulespinning, metal machining and jute processing, has been associated strongly and consistently with the occurrence of squamous-cell cancers of the skin, and especially of the scrotum. Oil and gas field work was reported in the past to be associated with acute myelogenous leukaemia, but this was not found in more recent studies. An increased risk of renal adenocarcinomas was seen for refinery and petrochemical workers and from occupational exposures to gasoline.

Environmental exposures have been reported to increase bone, brain and bladder cancer deaths of children and adolescents living in a residential area near three large petroleum and petrochemical complexes. Dermal carcinogenic potential of petroleum HCs was demonstrated by an increased incidence of squamous cell carcinomas and fibrocarcinomas in male mice treated with heavy paraffin distillate in lifetime skin painting studies. The limited data available on dermal absorption of PAHs do suggest that these compounds are rather well absorbed via the skin of humans as well as experimental animals (ATSDR, 1995b); absorption is facilitated if PAHs are present in a solvent, or in an oily or fatty vehicle. According to McKone and Howd (1992), absorption of an organic chemical through the skin is dependent on its lipophilicity, and decreases with increasing amounts applied on the skin. In addition, the actual absorption is also dependent on the matrix.

IARC classified the carcinogenic risk of residual fuel oils in category 2B: possibly carcinogenic to humans, based on sufficient evidence for its carcinogenicity in experimental animals and inadequate evidence for its carcinogenicity in humans (IARC, 1987).

PAHs are considered to be important contributors to the carcinogenic properties of heavy fuel oils, and have been extensively investigated with respect to their carcinogenicity. For evaluating the carcinogenic potency of PAH mixtures, the potency of the PAH relative to that of benzo[*a*]pyrene is estimated, which results in a benzo[*a*]pyrene equivalent. The key assumption is that the relative potency of two PAHs in an animal model is similar to that of the same components in humans. Furthermore the approach is based on the assumption of additivity, i.e. in the final risk estimation the individual PAH concentrations are multiplied with their respective potency factors, and the resulting benzo[*a*]pyrene equivalents are added up. A number of estimations for such carcinogenic potency factors have been made and have been evaluated recently by IPCS (1998). On the basis of this IPCS evaluation, RIVM concluded to potency factors as listed by Baars et al. (2001), expressed as the relative potency compared to the potency of benzo[*a*]pyrene which is defined as 1.00.

4. Limit values

4.1. General

In the assessment of the health risk for people involved in activities on the contaminated beaches, two types of risk associated with fuel oil # 6 components have to be considered: the risk due to toxicity and the risk due to carcinogenicity. In assessing toxic risks the actual exposure levels are compared with limit values taken from the literature; in assessing carcinogenic risks the actual exposure levels are compared with the $1{:}10^4$ lifetime excess risk for developing tumours.

4.2. Inhalation exposure

Regarding oil in general, ATSDR (1995b) lists a number of values for occupational exposure limits (TWA, time weight average) for mineral oil mists as established in a number of countries varying from 3 to 5 mg/m^3 (Australia, Belgium, former German Democratic Republic, Italy, The Netherlands, Switzerland, Japan, Finland, Sweden, USA).

Regarding benzo[*a*]pyrene, WHO estimated in its Air Quality Guidelines for Europe a unit lifetime risk of respiratory cancer of 8.7×10^{-5} per ng benzo[*a*]pyrene/m^3, which equals a lifetime

excess respiratory cancer risk of $1:10^4$ of 1.15 ng benzo[*a*]pyrene per m^3 (WHO, 1987). This value is based on the US-EPA evaluation of 1984, which in turn is based on occupational health studies of coke plant workers in the mid-1970s (US-EPA, 1984).

Regarding naphthalene, ATSDR (1995c) estimated a chronic inhalation MRL of 0.002 ppm which equals 10.5 μg/m^3; US-EPA estimated in 1998 a chronic inhalation RfC of 3 μg/m^3 (IRIS, 2000); the latter value has been used for the current risk assessment.

Regarding benzene, RIVM estimated the inhalation $1:10^4$ lifetime excess cancer risk to be 20 μg/m^3; regarding toluene a TCA of 400 μg/m^3 was derived, while for the xylenes a TCA of 870 μg/m^3 was derived (Baars et al., 2001).

4.3. Dermal exposure

Limit values for toxic risks due to dermal exposure to oil in general or to heavy fuel oils in particular are not available. CONCAWE (1998) reported significant skin irritation with rabbits dermally exposed to 1, 2 or 2.5 ml heavy fuel oil # 6 per kg bw per day (5 days treatment, 2 days rest, 5 additional days treatment). The dose of 1 ml/kg bw/day can be considered a LOAEL, and is equivalent to approximately 12 mg/cm^2/day. Since in general rabbit skin is more sensitive to chemical injuries than human skin, an uncertainty factor of 10 is applied to the LOAEL to derive a NOAEL of 1 mg/cm^2/day (rounded value), which is taken as the limit value in this particular exposure event without further adjustment.

For dermal exposure to PAHs in soil matrices RIVM estimated in 1989 a limit value for the $1:10^4$ excess lifetime skin cancer risk of 2 ng benzo[*a*]pyrene equivalents per cm^2 skin per day (Brinkman et al., 1989). This value was derived from animal skin painting experiments which had resulted in a limit value for the $1:10^4$ excess lifetime skin cancer risk of 0.3 ng benzo[*a*]pyrene (dissolved in acetone) per cm^2 skin per day. In deriving the limit value for PAHs in soil a correction was applied for dermal absorption of PAHs from soil, which was assumed to be approximately 15% of the dermal uptake from an acetone solution (McKone, 1990).

Data on dermal absorption during swimming are not available. However, assuming a flux of PAHs from water into skin similar to the flux from acetone into skin (McKone and Howd, 1992), results in a limit value for the $1:10^4$ excess lifetime skin cancer risk from exposure to PAHs (expressed as benzo[*a*]pyrene equivalents) in seawater of 125 ng/l[6]).

4.4. Oral exposure

A limit value for oral exposure to PAHs was derived by RIVM in 1999 on the basis of an extensive oral cancer risk study with rats administered benzo[*a*]pyrene, which resulted in a lifetime excess oral cancer risk of $1:10^4$ at an oral exposure of 0.5 μg benzo[*a*]pyrene per kg bw per day (Kroese et al., 1999; Baars et al., 2001).

5. Exposure assessment

5.1. General

In cleaning an oil spillage the exposure routes of consideration are in general the inhalation and the dermal route; oral exposure is prevented by appropriate safety measures and safety instructions.

Estimation of the potential inhalation exposure in cases of oil spillage is very difficult due to the general lack of data and the highly varying composition of heavy fuel oils in general. However, due to the low volatility of fuel oils, human

[6] According to McKone and Howd (1992), the flux of a lipophilic organic compound from an aqueous solution in contact with skin through the stratum corneum can be calculated as $J = K_p \times C_0$ (in which J is the flux in ng/cm^2/h, K_p is the skin permeability of the solution/skin system in cm/h, and C_0 is the concentration of the compound in ng/cm^3). McKone and Howd (1992) showed that for compounds with log $K_{ow} >$ 3 (as for most PAHs) the K_p tends to level at a value of 0.1 cm/h (or 2.4 cm/day). For the daily dermally applied dose of 0.3 ng benzo[*a*]pyrene per cm^2 as the limit value of the $1:10^4$ excess lifetime skin cancer risk, a flux of at least 0.3 ng/cm^2 per 24 h is assumed. With a K_p of 0.1 cm/h this flux is obtained at continuous dermal contact with a solution of 0.125 ng benzo[*a*]pyrene per cm^3 (equal to 125 ng/l) of which the concentration is kept constant.

exposure to vapour concentrations of toxicological significance is unlikely (ATSDR, 1995a). In exceptional situations, however, higher exposures seem possible, particularly if circumstances facilitate aerosol formation (IARC, 1989; ATSDR, 1995a).

Dermal exposure may result from contact with oil remnants and from contact with oil-contaminated seawater.

For tourists, an additional route of exposure is the accidental intake of seawater during swimming.

5.2. Beach and bird cleaning

People cleaning the beaches were assumed to be active for 8 h per day during 5 days per week. They wore full protective clothing, and thus the only exposure of interest is the inhalation route. People involved were assumed to be exposed to the atmosphere as shown in Table 4 (taking the highest levels of the two samples), with a risk for general toxicity due to exposure to volatile oil vapours including naphthalene, toluene and xylenes, and risk for carcinogenicity due to exposure to benzene (Table 6 part 1).

People cleaning the contaminated clothing using high pressure water devices also wore protective clothing, and thus again the oral route of exposure is irrelevant. This type of work was assumed to be done 1 h per day during 5 days per week. People involved were assumed to be exposed to an aerosol-like atmosphere similar to the experimentally generated atmosphere shown in Table 5, sample 2 (being the sample with the highest levels), with a risk for general toxicity due to naphthalene and a risk for carcinogenicity due to PAHs (Table 6 part 2).

Although the people who cleaned contaminated birds also wore protective clothing, the actual work was done with bare hands, and thus the exposures of concern are the inhalation route and the dermal route. This type of work was assumed to be done 8 h per day during 5 days per week. Next to inhalatory exposure to an atmosphere as shown in Table 4 (highest levels, with similar risks as outlined for beach cleaning people), their exposure involved dermal exposure to oil with a risk

Table 6
Exposure assessment

6.1. People cleaning beaches

Compound	Inhalation exposure: 8 h/d, 5 d/wk	Risk
Volatile organic compounds	4450 μg/m^3	General toxicity
Benzene	124 μg/m^3	Carcinogenicity
Toluene	576 μg/m^3	General toxicity
Xylenes	1995 μg/m^3	General toxicity
Naphthalene	4.6 μg/m^3	General toxicity

6.2. People cleaning clothes

Compound	Inhalation exposure: 1 h/d, 5 d/wk	Risk
Benzo[*a*]pyrene (equivalents)	33 ng/m^3	Carcinogenicity
Naphthalene	680 ng/m^3	General toxicity

6.3. People cleaning birds

Compound	Dermal exposure: 8 h/d, 5 d/wk	Risk
Oil	11.6 mg/cm^2	General toxicity
Benzo[*a*]pyrene equivalents	1240 ng/cm^2	Carcinogenicity

6.4. Tourists, dermal exposure on the beach

Compound	Dermal exposure: 1 h/day	Risk
Benzo[*a*]pyrene equivalents	10 ng/cm^2	Carcinogenicity (children and adults)

6.5. Tourists, dermal exposure during swimming

Compound	Dermal exposure: 4 h/day	Risk
Benzo[*a*]pyrene equivalents	10 ng/l	Carcinogenicity (children and adults)

6.6. Tourists, ingestion of water during swimming

Compound	Oral exposure: ng/kg bw/day	Risk
Benzo[*a*]pyrene equivalents	1.0	Carcinogenicity, children
Benzo[*a*]pyrene equivalents	0.07	Carcinogenicity, adults

for general toxicity due to contact with oil and a carcinogenicity risk due to contact with PAHs in this oil. It was assumed that dermal exposure of oil occurred on both hands, 8 h/day, 5 days/week, with at average about 10 g of oil. Taking for both hands together a skin surface of 860 cm^2 (Bremmer and Van Veen, 2000), this resulted in an exposure of 11.6 mg oil per cm^2 unprotected skin. Taking beach sample 2 as listed in Table 3 as representative for the oil to which people cleaning birds were dermally exposed led to a total dermal exposure of 1240 ng benzo[*a*]pyrene equivalents per cm^2 per 8 h working day (Table 6 part 3).

5.3. Tourist activities

With regard to general tourist activities on the beaches, this is interpreted to concern people spending time on the beach with general leisure activities like relaxing, sunbathing, swimming, playing around, etc. A typical 'day at the beach' is defined as a period of 10 h at most, with sea swimming for at most 4 h per day. At the start of the holiday season (June 2000), about 5 months had elapsed since the wreckage of the Erika and the pollution of the shore. By that time the tourist beaches were largely cleaned, and thus the risk due to inhalation exposure was considered negligible. Furthermore, if any oil remnants would be present, it is to be expected that parents and guardians will closely observe their children and pupils to avoid accidental oral intake of these, and thus the health risk following oral exposure by intake of soil is also considered negligible.

People staying on the beach were at risk of dermal exposure due to accidental skin contact with oil remnants on the beach, which is assumed to have occurred with about 25 g of tar-like oil remnants sticking to the sole of the foot, for 1 h per day at most (because if such a contact should occur, it is likely that it will be noticed and removed as far as possible and as soon as possible). In this exposure the risk of concern is the development of skin tumours due to the PAHs present in these oil remnants. Assuming for the soles of both feet together a skin surface of 500 cm^2 (derived from the skin surface of both feet which is 1170 cm^2 (Bremmer and Van Veen, 2000)), this results in 50 mg oil per cm^2 skin. The data regarding the PAH content of the oil on the beach (see Table 3) suggest a rather rapid decrease of this content due to weathering: the half-life of the decrease appears to be (conservatively estimated and assuming a first-order process) approximately 1 week. This would result in an actual PAH content of the oil remnants (May/June 2000, assuming an average stay of such oil remnants on the beach of 10 weeks) of about 0.1% of the original content of approximately 200 mg benzo[*a*]pyrene equivalents per kg fresh oil. Thus 50 mg of weathered oil remnants per cm^2 skin would contain $50 \times 0.2 = 10$ ng benzo[*a*]pyrene equivalents per cm^2 skin, for a period of 1 h per day (Table 6 part 4).

Furthermore, tourists were dermally exposed during swimming (4 h per day at most) in seawater that possibly contains oil, because oil may still be leaking from the wreck. The actual risk of concern is the development of skin tumours resulting from exposure to PAHs in dissolved oil. The aqueous solubility of fuel oil # 6 is 5 mg/l at most (see Table 1). Due to the fact that the distance between the wreck and the nearest coast is approximately 65 km, and taking into account the strong local currents and tidal movements, it is expected that near the beaches the dissolved oil will be diluted with a factor of 100 at least. This results in a maximum oil concentration at beach level of 50 μg/l, and thus the local seawater may contain 10 ng benzo[*a*]pyrene equivalents per l (cf. Table 3, rounded value for fresh oil). Thus, at swimming for 4 h/day the body was in full contact with water containing 10 ng benzo[*a*]pyrene equivalents per l at most (Table 6 part 5).

Finally, during swimming, tourists were orally exposed to seawater containing dissolved PAHs, with a risk for carcinogenicity. For adults an ingestion of 0.5 l seawater during a full day at the beach (4 h swimming) is assumed. This amount contained 25 μg oil at most, containing 5 ng benzo[*a*]pyrene equivalents. For an adult with a body weight of 70 kg this equals a (daily) dose of $5/70 = 0.07$ ng/kg bw/day. For children an ingestion of 1 l seawater during a full day at the beach (4 h swimming) is assumed. This amount con-

tained 50 μg oil, containing 10 ng benzo[*a*]pyrene equivalents. For a small child with a body weight of 10 kg this equals a (daily) dose of $10/10 = 1$ ng/kg bw/day (Table 6 part 6).

6. Risk characterisation

6.1. Beach and bird cleaning

For people cleaning the beaches the actual inhalation after correction with a factor of $(8/24) \times (5/7)$ for continuous exposure time during at least a full week (except the inhalation of volatile organic compounds because this particular exposure is compared with a limit value for occupational health) is for the larger part well below the limit values (Table 7 part 1). For benzene (which is carcinogenic), however, the corrected inhalation for continuous exposure time during at least a full week (29.5 μg/m^3) is slightly higher than the limit value (20 μg/m^3), but taking into account that the period during which these people are exposed is very short (generally in the order of magnitude of some weeks to some months at most) compared to the limit value which is based on continuous exposure during full lifetime (assumption: 4 months versus 70×12 months), the risk can also be considered negligible (Table 7 part 1).

In comparing the exposure to PAHs of people cleaning clothes with the limit value it must be noted that the WHO Air Quality Guideline of 1.15 ng/m^3 is valid for benzo[*a*]pyrene solely serving as an index for general PAH mixtures (WHO, 1987), while the actual exposure level is expressed in benzo[*a*]pyrene equivalents. Taking the ratio of *benzo[a]pyrene:benzo[a]pyrene equivalents* in the actual oil spillage to be at average 1:1.5 (Table 3), this results in an actual exposure after correction with a factor of $(1/24) \times (5/7)$ for continuous exposure during at least a full week of 0.65 ng benzo[*a*]pyrene per m^3, to be compared with the limit value of 1.15 benzo[*a*]pyrene per m^3. As shown in Table 7 part 2, the risk for these people can be considered to be negligible, both with regard to general toxic effects due to naphthalene exposure (corrected exposure level is 20 ng/m^3, the limit value is 3000 ng/m^3), and with regard to inhalation carcinogenicity due to exposure to benzo[*a*]pyrene and other carcinogenic PAHs.

For people cleaning birds the dermal exposure to oil (a time-corrected full day exposure level of 4 mg/cm^2, while the limit value is estimated to be approximately 1 mg/cm^2) may well lead to a serious risk of rather acutely developing skin disorders like irritant and defatting effects, resulting in primary irritation, dryness, cracking, oil acne, dermatitis and/or hyperkeratosis (Table 7 part 3). In addition, the dermal exposure to this oil may give rise to sensitisation and to some eye irritation. These risks, however, are very difficult to quantify. Generally the effects are reversible, although proper medical treatment might be required.

Apparently the calculated risk for developing skin tumours at some time in the future is considerable, also after correction with a factor of $(8/24) \times (5/7)$ for continuous exposure during at least a full week (which resulted in an exposure level of almost 300 ng/cm^2, while the limit value is 2 ng/cm^2; Table 7 part 3). However, again it must be realised that the actual exposure period is at most in the order of magnitude of 1 month, compared to the limit value which is based on continuous exposure during full lifetime (in other words, approximately one month versus 70×12 months). In conclusion, and given the actual situation, the risk may well considered to be acceptable.

6.2. Tourist activities

For tourists the dermal exposure due to skin contact with oil remnants on the beach for 1 h per day is equivalent with an exposure of 0.4 ng/cm^2 after correction with a factor of 1/24 for continuous exposure during at least a full day. Compared with the limit value of 2 ng/cm^2 the actual risk resulting from this exposure is considered to be negligible (Table 7 part 4).

Swimming tourists are dermally exposed to seawater estimated to contain 10 ng benzo[*a*]pyrene equivalents per l, during 4 h per day at most. Corrected for continuous exposure during at least a full day (applying a factor of 4/24) this results in an exposure level of 1.7 ng benzo[*a*]pyrene equiv-

Table 7
Risk characterisation

7.1. People cleaning beaches

Compound	Risk	Corrected inhalation exposure (continuously, 24 h/d, 7 d/wk)	Limit value
Volatile organic compounds	General toxicity	4450 μg/m^3[a]	3000–5000 μg/m^3
Benzene	Carcinogenicity	29.5 μg/m^3	20 μg/m^3[b]
Toluene	General toxicity	137 μg/m^3	400 μg/m^3
Xylenes	General toxicity	475 μg/m^3	870 μg/m^3
Naphthalene	General toxicity	1.1 μg/m^3	3.0 μg/m^3

7.2. People cleaning clothes

Compound	Risk	Corrected inhalation exposure (continuously, 24 h/d, 7 d/wk)	Limit value
Benzo[*a*]pyrene (equivalents)	Carcinogenicity	0.65 ng/m^3[c] 0.98 ng/m^3[d]	1.15 ng/m^3[b,c]
Naphthalene	General toxicity	20 ng/m^3	3000 ng/m^3

7.3. People cleaning birds

Compound	Risk	Corrected dermal exposure (continuously, 24 h/d, 7 d/wk)	Limit value
Oil	General toxicity	4 mg/cm^2[e]	1 mg/cm^2
Benzo[*a*]pyrene equivalents	Carcinogenicity	295 ng/cm^2	2 ng/cm^2[f]

7.4. Tourists, dermal exposure on the beach

Compound	Risk	Corrected dermal exposure (continuously, 24 h/day)	Limit value
Benzo[*a*]pyrene equivalents	Carcinogenicity (children and adults)	0.4 ng/cm^2	2 ng/cm^2[f]

7.5. Tourists, dermal exposure during swimming

Compound	Risk	Corrected dermal exposure (continuously, 24 h/day)	Limit value
Benzo[*a*]pyrene equivalents	Carcinogenicity (children and adults)	1.7 ng/l	125 ng/l[f]

7.6. Tourists, ingestion of water during swimming

Compound	Risk	Oral exposure	Limit value
Benzo[*a*]pyrene equivalents	Carcinogenicity, children	1.0 ng/kg bw/day	500 ng/kg bw/day[g]
Benzo[*a*]pyrene equivalents	Carcinogenicity, adults	0.07 ng/kg bw/day	500 ng/kg bw/day[g]

[a] Not corrected for 7 days continuous exposure since the limit value is an occupational health limit based on exposure for 8 h/day, 5 days/week.
[b] 1:10^4 excess lifetime cancer risk from inhalation exposure.
[c] Expressed as benzo[*a*]pyrene solely, see text.
[d] Expressed as benzo[*a*]pyrene equivalents, see text.
[e] Only corrected for 24 h exposure since the limit value was directly derived from a rabbit experiment in which the animals were dermally exposed for two periods of 5 days separated by a 2 days rest period.
[f] 1:10^4 excess lifetime cancer risk from dermal exposure.
[g] 1:10^4 excess lifetime cancer risk from oral exposure.

alents per l. Compared with the estimated limit value of 125 ng/l, the actual risk is considered to be negligible (Table 7 part 5).

In addition, swimming tourists are also orally exposed to this seawater via accidental swallowing of some water. The estimated intakes from this exposure, 1.0 and 0.07 ng benzo[*a*]pyrene per kg bw/day for children and adults, respectively, are very small compared to the limit value of 500 ng/kg bw/day (Table 7 part 6). Hence also these risks are considered to be negligible for both children and adults.

7. Conclusion

At first instance the health risks for people (among which many volunteers) cleaning the beaches and the birds immediately following the fuel oil # 6 spillage resulting from the wreckage of the 'Erika' appeared to be considerable. Hence there was an urgent need for an evaluation of these risks to allow the local and national authorities to take proper measures. The risk estimation, however, was hampered by (1) a general lack of detailed chemical–analytical and exposure data of this particular oil spillage, (2) very scarce data with respect to exposure–effect relationships regarding heavy fuel oils, and (3) only a few reliable limit values for the different oil components. Consequently, considerable assumptions (some of them rather worst-case) regarding the various exposure scenarios had to be made.

Bearing in mind these limitations, the actual risk assessment resulted in generally negligible risks both for people involved in the various cleaning operations as well as for tourists spending leisure time on the beaches some months later. The one exception appeared to be the people cleaning oil-contaminated birds with bare hands: they were at risk for developing (sub)acute eye and skin irritation and dermatitis, and at risk for developing skin tumours at some time in the future. Eye and skin irritation, however, are in general reversible adverse effects, and thus the risk for permanent damage is negligible. The risk for developing skin tumours was considered small in view of the limited exposure time compared with the limit value, which is based on life-long continuous exposure.

7.1. Remark

It must be noted that in the above evaluation the normal background exposure, to which the average human is exposed in normal daily life, is not taken into account. In general, however, the background exposure to the compounds of interest in this particular oil spillage (consisting of heavy fuel oil # 6) can be expected to be low.

With respect to PAHs, the background exposure of the general population is approximately 240 ng PAHs (corresponding to 7.3 ng benzo[*a*]pyrene equivalents) per kg bw per day via the oral, and about 6.5 ng PAHs (corresponding to 0.9 ng benzo[*a*]pyrene equivalents) per kg bw per day via the inhalatory route of exposure (IPCS, 1998; Baars et al., 2001). Smokers have a considerably higher exposure by inhalation.

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References

ATSDR, 1995a. Toxicological profile for fuel oils. US Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA.

ATSDR, 1995b. Toxicological profile for polycyclic aromatic hydrocarbons (update). US Department of Health and

Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA.

ATSDR, 1995c. Toxicological profile for naphthalene (update). US Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA.

ATSDR, 1999. Toxicological profile for total petroleum hydrocarbons. US Department of Health and Human Services, Public Health Service, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA.

Baars, A.J., Theelen, R.M.C., Janssen, P.J.C.M., Hesse, J.M., Van Apeldoorn, M.E., Meijerink, M.C.M., Verdam, L., Zeilmaker, M.J., 2001. Re-evaluation of human-toxicological maximum permissible risk levels. RIVM report no. 711701025, March 2001. National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

Boudet, C., Chemin, F., Bois, F., 2000. Evaluation du risque sanitaire de la marée noire consécutive au naufrage de l'Erika. Partie 6. Institut National de l'Environnement Industriel et des Risques (INERIS), report CBoFChFBo-25627 dated February 29, 2000. Verneuil-en-Halatte, France.

Bremmer, H.J., Van Veen, M.P., 2000. Factsheet algemeen—randvoorwaarden en betrouwbaarheid, ventilatie, kamergrootte, lichaamsoppervlak (General factsheet—limiting conditions and reliability, air change rate and volume of Dutch dwellings, body surface. Report in Dutch). RIVM report no. 612810009, March 2000. National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

Brinkman, F.J.J., Knaap, A.G.A.C., Kramers, P.G.N., Aalbers, Th.G., Jekel, A.A., Keijzer, J., Kliest, J.J.G., Michel, F.J., Montizaan, G.K., Savelkoul, T.J.F., Vaessen, H.A.M.G., Wammes, J.I.J., Wilbers, A.A.M.M., 1989. Onderzoek naar de gehalten aan polycyclische aromaten in binnen het voormalige Laura-terrein te Kerkrade verzamelde monsters. Uitloogbaarheidsproeven. Risico-evaluatie met betrekking tot de volksgezondheid. (Concentrations of polycyclic aromatic hydrocarbons in samples collected at the former Laura coal-mine area at Kerkrade. Leaching experiments. Risk evaluation with respect to public health. Report in Dutch). RIVM report no. 748704018, April 1989. National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

CONCAWE (the Oil Companies' Organisation for Environmental and Health Protection), 1998. Heavy fuel oils. Product Dossier No. 98/109. CONCAWE, Brussels, Belgium.

IARC, 1987. Overall evaluations of carcinogenicity—an updating of IARC monographs volumes 1 to 42. International Agency for Research on Cancer, Monographs on the Evaluation of Carcinogenic Risks to Humans, Suppl. 7. IARC, Lyon, France.

IARC, 1989. Occupational exposures in petroleum refining; crude oil and major petroleum fuels. International Agency for Research on Cancer, Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 45. IARC, Lyon, France.

IPCS (International Programme on Chemical Safety), 1982. Selected petroleum products. Environmental Health Criteria 20. WHO/IPCS, Geneva, Switzerland.

IPCS (International Programme on Chemical Safety), 1998. Selected non-heterocyclic polycyclic aromatic hydrocarbons. Environmental Health Criteria 202. WHO/IPCS, Geneva, Switzerland.

IRIS (Integrated Risk Information System), 2000. Summary for naphthalene (dated September 1998). US Environmental Protection Agency, Washington, DC, USA.

Irwin, R.J., VanMourik, M., Stevens, L., Seese, M.D., Basham, W., 1997. Environmental Contaminants Encyclopaedia-Entry on Fuel Oil No. 6. National Park Service, Water Resources Division, Fort Collins, CO, USA. Distributed on the Internet via the US National Park Service Home Page: http://www.aqd.nps.gov/toxic/.

Kroese, E.D., Muller, J.J.A., Mohn, G.R., Dortant, P.M., Wester, P.W., 1999. Tumorigenic effects in Wistar rats orally administered benzo[a]pyrene for two years (gavage studies). Implications for human cancer risks associated with oral exposure to polycyclic aromatic hydrocarbons. RIVM report no. 658603010 (in preparation), draft of July 1999. National Institute of Public Health and the Environment, Bilthoven, The Netherlands.

McKone, T.E., 1990. Dermal uptake of organic chemicals from a soil matrix. Risk Anal. 10, 407–419.

McKone, T.E., Howd, R.A., 1992. Estimating dermal uptake of non-ionic organic chemicals from water and soil. I. Unified fugacity-based models for risk assessments. Risk Anal. 12, 543–557.

TPHCWG (Total Petroleum Hydrocarbons Criteria Working Group, Toxicology Technical Action Group), 1997a. Selection of representative TPH fractions based on fate and transport considerations. TPHCWG Series Volume 3. Amherst Scientific Publishers, Amherst, MA, USA.

TPHCWG (Total Petroleum Hydrocarbons Criteria Working Group, Toxicology Technical Action Group), 1997b. Development of fraction specific reference doses (RfDs) and reference concentrations (RfCs) for total petroleum hydrocarbons. TPHCWG Series Volume 4. Amherst Scientific Publishers, Amherst, MA, USA.

US-EPA, 1984. Carcinogen assessment of coke oven emissions. US Environmental Protection Agency, Final report no. EPA-600/6-82-003F. Washington, DC, USA (cited in WHO 1987).

WHO, 1987. Air quality guidelines for Europe. World Health Organization, Regional Office for Europe, WHO Regional Publications, European Series No. 23. Copenhagen, Denmark.